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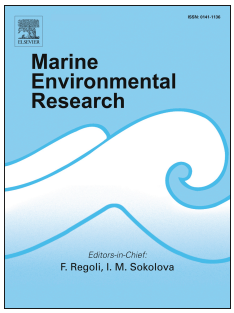

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Comparison of caged and native blue mussels (*Mytilus edulis* spp.) for environmental monitoring of PAH, PCB and trace metals

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Abstract

Contaminant bioaccumulation was studied in blue mussels (*Mytilus edulis* spp.) using the harbor waters of Kristiansand (Norway) as a case study. A suite of chemical contaminants (trace metals, PAHs and PCBs) was analyzed in caged and native mussels as well as in passive samplers (Diffusive Gradients in Thin films (DGT)-devices and silicone rubbers) placed alongside the mussels for estimation of contaminant concentrations in water and uptake rates and bioaccumulation factors (BAFs) in mussels during a six-months deployment period. Estimated logBAFs were in the ranges 2.3 - 5.5, 3.8 - 5.2 and 3.2 – 4.4 for metals, PCBs and PAHs, respectively. Contaminant levels in caged mussels increased rapidly to stable levels for trace metals, whereas for hydrophobic organic contaminants the increase was steady but slow and for many compounds did not reach the levels observed in native mussels. Some key issues related to mussel caging design, such as mussel deployment time and confounding influence from seasonal fluctuations, are discussed herein.

Keywords: blue mussels; biomonitoring; caging; contaminant bioaccumulation factors

1. Introduction

Blue mussels (*Mytilus* spp.) are widely used as sentinels in coastal pollution monitoring (mussel watch) programs, mainly because their biological characteristics make them very suitable as bioindicators for assessing the quality status of coastal waters (Farrington et al., 2016; Beyer et al., this volume). Most often mussel watch studies involve collection of samples from natural blue mussel populations, but the adoption of an active biomonitoring alternative by using transplanted blue mussel has gained considerable popularity in ecotoxicology research and monitoring. Indeed, the straightforwardness of using controlled deployments is one of the key advantages with blue mussels in

marine monitoring. The comparability of deployed and native mussels in pollution biomonitoring has therefore been investigated in a number of field studies, e.g. (Regoli and Principato, 1995; Peven et al., 1996; Walsh and O'Halloran, 1998; Nasci et al., 2002; Ericson et al., 2002; Nigro et al., 2006), and others have suggested that an integrated use of monitoring data from both native and transplanted mussels may provide a more accurate assessment of pollutant uptake and effect phenomena at contaminated field locations, e.g. (Bodin et al., 2004; Bebianno et al., 2007; Serafim et al., 2011; Brooks et al., 2012). A key question for all such caging studies is how long the blue mussels should stay deployed to be fully representative as a biological sample for assessment of pollutant concentrations and other ecotoxicological parameters.

Mussel caging is particularly useful when indigenous mussels are scarce or absent at the planned study sites. The mussel caging alternative is therefore increasingly more being used in trend monitoring (spatial and temporal) and in industrial compliance monitoring (e.g. comparing to quality standards or regulatory benchmarks). However, the actual comparability of caged and native mussels under the specific study conditions is often insufficiently documented. It may for example be relevant to clarify how key factors such as deployment time, caging design (e.g. fixed or floating mussel rig setups), genetic homogeneity/variability of the caged mussels, etc., could affect the general outcome of the study. The investigator may often want to manipulate key study factors (such as timing and duration of exposure, positioning of the caged specimens, etc.) in a controlled manner to create more accurate study designs and to increase the overall quality of the monitoring data. In Norway, technical requirements for mussel monitoring are embedded in national environmental regulations which recently were updated to comply with demands expressed in the EU Water Framework Directive (WFD, 2000/60/EC) and the Marine Strategy Framework Directive (MSFD, 2008/56/EC). These are two wide-ranging trans-national environmental legislation frameworks designed for the protection and restoration of aquatic environments in Europe, see Borja et al. (2010). Relevant requirements relate to representative positioning of stations, choice of sample matrices and the use of quality standards (QSs) for evaluation of quality status based on contaminant concentration data. With this in mind, a further harmonization of the concept of mussel caging could be important, as it may facilitate the standardization of field monitoring designs and better comparability of coastal monitoring conducted in different countries.

In this study, we study contaminant bioaccumulation in blue mussels transplanted to the waters of the city harbor of Kristiansand (Norway), an area known to be moderately to severely polluted by a mixture of inorganic and organic contaminants; especially nickel, copper, cobalt, polycyclic aromatic hydrocarbons (PAHs) and hexachlorobenzene (HCB). The caged mussels, and also native mussels from the harbor, were repeatedly sampled during a period of six months and analyzed for trace metals, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), and chlorinated

benzenes. Co-deployed passive sampling devices (Diffusive Gradients in Thin-films (DGTs) and silicone rubbers) were used to estimate freely dissolved contaminant concentrations in the seawater and this allowed the calculation of uptake and excretion rates as well as bioaccumulation factors (BAFs) of contaminants in mussels based on first-order single-compartment toxicokinetics. The results of the present study are relevant in the context of an ongoing work coordinated by the Norwegian Environmental Agency (NEA) and Standard Norway (SN) aiming to develop a Norwegian Standard (NS) for how to use blue mussels and blue mussel caging in marine pollution monitoring.

2. Material and methods

2.1 Study design and field work

The field work was conducted in the period late May – late November 2015. A suitable number of blue mussels (size range 3-5 cm) was obtained from a mussel farm located in Kaldvellfjord (Lillesand, Norway), a locality distant from known point sources of contamination. First, two replicate samples each including 60 individual mussels were grouped and frozen to serve as before-deployment controls for the caged mussels. The other mussels were transported rapidly (in a cooling box equipped with cooling elements and some brown algae to keep humid conditions) to the caging stations in the Kristiansand city harbor area. The mussels were out of water for only about two hours. Caging Stations 1 and 2 were located in the inner and outer part of the harbor, respectively (Figure 1). Station 1 (GPS position 58.13713, 7.97239) was located by the quay of a metal processing plant that produces high quality Ni as well as Co and Cu, whereas Station 2 was located about 2 km in SSE direction from Station 1 in the outer harbor area by the small islet Svensholmen (58.12546, 7.9878).

Mussel caging rigs, based on collapsible 5-floor lantern nets (1 m vertical height), were prepared and equipped with approx. 1000 mussels per rig. Passive samplers (DGTs and silicone rubbers) were also mounted in duplicate at each rig. Field control samplers were used to assess contamination in unexposed samplers and in the case of silicone rubber samplers to measure initial performance reference compound concentrations. Samplers were all from the same batches and were all analyzed together. The rigs were positioned in the sea by means of buoys, ropes and weights at each station. The upper end of the lantern net was approx. at 2 m depth. Native mussels of suitable size were only living at caging Station 2 (Svensholmen). The mussel population at Station 2 had been monitored annually since 1998 in connection with the Norwegian contribution to the Oslo-Paris Commission (OSPAR) Joint Assessment and Monitoring programme (JAMP) (Green et al., 2016). After the start of mussel deployment at May 29th, 2015 (day 0) each rig was sampled after approximately one month (July 2nd, 34 days), three months (Sept. 10, 104 days) and six months (Nov. 26, 181 days). Samples of native mussels (at Station 2, Svensholmen) were collected at the start-up day (in late May) and

subsequently the same days as for the caged mussels. For each sampling day, approx. 150 mussels were retrieved from each rig and from the Svensholmen population and transported (cold and humid) to the NIVA laboratory (Grimstad) to be frozen and stored to sample preparation. The mussels were not depurated before freezing. Sea temperature data at caging stations were obtained at each sampling day. Unfortunately, at the last sampling, the mussel rig at Station 2 had disappeared for an unknown reason, thus these data (caged mussels and passive samples after six months at Station 2) are lacking in this study. The DGT samplers at both rigs were retrieved after one month of deployment to limit the impact of fouling developing at the surface of the sampler.

2.2 Sample preparation and chemical analyses

The mussel sample preparation was performed according to the OSPAR guidelines (OSPARCOM, 2012) and with further details described by Green et al. (2016). The frozen mussels were thawed and two replicate composite samples (each consisting of 60 mussels) per station and per sampling day were prepared for the transplanted and native mussels, respectively. The number of composite samples was decided based on cost-effectiveness. In general, the optimal number of composite samples and the number of individuals per sample depends on the cost of chemical analyses relative to sampling and sample preparation, as well as the level of inherent variation among individuals due to e.g. physiological factors (Bignert et al., 2014). The shells were scraped clean on the outside; the length was measured by means of slide calipers; all soft tissue was scraped out by using a scalpel, weighed, and merged to a composite sample which was weighed before it was frozen and stored at -20 °C until further homogenization and analysis. Empty shells were dried and then weighed sample-wise for condition index estimation. Each pooled mussel sample was analyzed for As, Cd, Cr, Cu, Hg, Ni, Pb, Zn, the 16 US EPA PAHs (EPA Methods 550.1/610/8100/8270C/8310), the 7 indicator PCBs (CB 28, 52, 101, 118, 138, 153 and 180), hexachlorobenzene, pentachlorobenzene, fats/lipids and dry matter by using the analytical methods described by Green et al. (2008). Quality Assurance (QA) of chemical analyses at NIVA and Eurofins are carried out by participation in international intercalibration exercises (QUASIMEME) and other relevant proficiency testing programs with acceptable results (Green et al., 2016). Certified reference materials (CRM), Standard Reference Materials (SRM) (e.g. DORM-4 fish protein and QUASIMEME reference biota samples) and in-house reference materials are analyzed routinely. The laboratories are accredited according to ISO/IEC 17025:2005. Chemical analyses were performed on wet tissue samples and the content of solids and lipid were measured to enable statistical examination of chemical concentration data at a wet weight (wet wt.), dry wt., and lipid wt. basis. The data reporting format is specified in table and figure legends. Freely dissolved contaminants concentrations (C_{free}) were estimated from passive samplers, DGTs for metals and silicone rubbers for PCBs and PAHs. The DGT passive samplers were analyzed for Al, Ca, Cd, Co, Cr, Cu, Fe, Ni, Pb and Zn (but not Hg), while the silicone rubber samplers were analyzed for the 16

US EPA PAHs, the 7 indicator PCBs, HCB and pentachlorobenzene following procedures described by Allan et al. (2013). The preparation, extraction and analysis procedures and data of DGT and silicone rubber passive samplers are shown in the Supporting Information. Field control passive samplers were used to estimate possible contaminant levels present in non-exposed samplers and in the case of silicone rubber passive samplers, the measurement of initial performance reference compound concentrations, as recommended by Booij et al. (2006) for silicone rubber and by Dabrin et al. (2016) for DGT passive samplers. The NIVA laboratory participated in QUASIMEME intercomparison exercises on passive sampling with AlteSil™ silicone rubber in 2014 and 2015 and obtained excellent results.

2.3 Treatment and statistical examination of data

Contaminant concentrations in caged and native mussels and in passive samplers were compared and examined for station-wise and temporal trends during the six months' study period. Single-compartment uptake/elimination modelling was employed for evaluating the contaminant bioaccumulation processes. Statistical analysis was performed with the use of R software (version 3.3.2) and Statistica software (version 7.1, StatSoft, Tulsa, OK, USA). Differences were evaluated using Analysis of Variance (ANOVA). The small sample size is itself not invalidating ANOVA so long as the assumptions are met. Checking the normality assumption, which is critical with a low sample size, is not feasible using graphical methods as there are only two samples per site/date/treatment. However, the use of composite samples of a large number of mussels has the effect of normalizing the data greatly. Thus, even when the distribution of concentrations in individual mussels is extremely skewed, the distribution of concentrations in a composite sample of 60 mussels is expected to be close to normal. Levene's test was used to test for heterogeneity of variance. When necessary, data were $\log_{10}$-transformed to reduce heterogeneity of variance. In some cases, both deployed and native mussels showed the same general and approximately linear trends over time; in these cases, we used ANCOVA to analyze the difference between deployed and native mussels across sampling occasions. A significance level of $\alpha = 0.05$ was chosen. Due to the low sample size, p-values should be interpreted with some caution when p-values are between 0.01 and 0.05; on the other hand, it should also be kept in mind that the statistical power is low. The regression tool in Sigmaplot was used to obtain BAFs and contaminant depuration rates (k_2) for contaminants accumulating in mussels. Modelling of the uptake of organic contaminants (PAHs and PCBs) in native mussels at the Svensholmen site was done by using Equation (1), as described by Björk and Gilek (1999):

$$C_{m,t} = C_{m,t_0} + C_{free}\left(BAF - \frac{C_{m,t_0}}{C_{free}}\right)\left(1 - e^{-k_2 t}\right) \quad \text{(Eq. 1)}$$

where C_m is the concentration in mussels (ng/kg wet wt.), C_{free} is the freely dissolved concentration from silicone rubbers (ng/L), BAF is in L/kg, and k_2 is the 1st order mussel depuration rate (d^{-1}). $C_{m,t0}$

and C_{free} are known, BAF and k_2 were estimated from the modelling of contaminant uptake at Station 1 (industrial harbor site) (when k_2 values were not obtained, we used the median of values reported for PAHs or PCBs), and t is either 34 or 104 d. The use of Equation 1 in relation to data obtained from passive samplers was performed as described by Booij et al. (2006).

3. Results

Biological data and the results of chemical contaminant measurements in 0-group reference, deployed and native mussels in the present study are shown in Table 1. The survival of deployed mussels during the six-months caging period was very good, with practically no mortality, but during the caging period a lowering trend of lipid content was recorded in caged mussels. This trend was also observed in native mussels, indicating seasonal fluctuations in the study area. However, this general decrease of lipid content would obviously have an influence on the accumulation of contaminants, and in particular of the hydrophobic substances.

The chemical analysis of the reference mussels from the donor site (Kaldvellfjord) confirmed generally low contaminant concentrations in the pre-deployed mussels (Table 1), except for Cu which was found to be approximately twice the level expected for an unpolluted sample. After being transplanted, a substantial increase of multiple contaminants was observed in caged mussels at both stations. The increase was most pronounced for nickel at Station 1 (Figure 2), which increased up to 35 times when compared with the concentration in pre-deployed mussels (Table 1). The DGT passive sampler accumulates labile metal species from solution while deployed *in situ*, thereby providing an estimate of the bioavailable fraction of metals; which will include both free metal ions and kinetically–labile metal complexes (i.e., those with rapid dissociation kinetics) (Zhang and Davison, 1995). The relationship between the concentrations of metals in blue mussels (both transplanted and native) and labile metal concentrations measured with the DGT sampler is shown in Figure 3. Based on the measurement of DGT-labile concentrations, blue mussel-water bioaccumulation factors for metals measured in transplanted and native mussels could be calculated (Table 2). Interestingly, as shown in Figure 4, the various metals detected in this study showed variable uptake patterns in transplanted and native mussels, e.g. with concentrations of Hg (not analyzed in DGTs), Pb, Fe and Cd being relatively higher in the native mussels than in the transplanted mussels whereas aluminum loads were generally higher in the transplanted samples.

The bioaccumulation curves for the different PCB congeners detected in caged mussels at Station 1 varied significantly based on the degree of chlorination (and thus hydrophobicity) (Figure 5). As shown in the results overview in Table 1, there was a noticeable difference in PCB concentration levels between the pre-deployment mussel sample (which showed the lowest levels) and all other mussel samples (both deployed and native), and the native mussels at Svensholmen displayed a

slightly higher PCB level than the deployed mussels at all time points (for PCB 7, $t = 6.45$, $p < 0.001$ in ANCOVA with time). Lipid-normalized concentrations of CB 138 and 101 showed a highly linear increase over time for transplanted mussels at both sites (CB 101: $t = 3.11$, $p = 0.036$; CB 138: $t = 5.04$, $p < 0.01$) (Figure 6). Some of the less chlorinated congeners (i.e. CB 28 and 31), however, displayed a very different pattern, namely by increasing sharply during the first month before decreasing during the following months. CB 101 and 118 showed an intermediate pattern by apparently plateauing after 50 days of deployment at Svensholmen and after 100 days at the industrial harbor (Figure 6). In the native mussels at Svensholmen, the PCB levels showed a slight but not significant increase ($p > 0.2$) during the six months' study period, as shown in Table 1.

For PAHs, the highest concentrations were found in the native mussel samples (at Station 2, Svensholmen), especially at the last sampling day towards the end of the study period (Table 1). Unfortunately, at this last sampling point, the caging rig at the Svensholmen site had disappeared. However, the key trend in native mussels was that all PAH concentrations decreased from May to July, and then increased again to September and even more to November (Table 1, Figure 7). This was most likely related to spawning and a resulting loss of tissue lipids in the early deployment period. For PAHs in deployed mussels, there was a clear increase in concentration between pre-deployed to the deployed groups (similar as for the PCBs), emphasizing the non-polluted nature of the donor population at the Kaldvellfjord site. The two caging groups were slightly different, and interestingly, Station 2 site exhibited higher concentrations than Station 1 ($t = 4.14$, $p < 0.01$; Table 1), i.e. an opposite pattern to that seen for metals. The main uptake patterns of PAHs in caged mussels were relatively similar at the two sites and concentrations of several PAHs (as pyrene, fluorene and chrysene) increased linearly on lipid wt. basis at both sites (Figure 7). A comparison of the Station 2 native mussels to the pre-deployed mussels clearly show that the waters at Station 2 were quite markedly contaminated with PAHs, with concentrations of some PAHs up to >50 times higher for Station 2 native mussels (Table 1).

BAF values (wet wt.) for PAHs and PCBs in blue mussels are shown as a function of $\log K_{ow}$ in Figure 8, and the estimated BAF values for the different PCBs and PAHs are shown in Table 3 and Table 4, respectively. In Figure 8, data are plotted against literature-based $\log K_{ow}$-logBAF relationships from Booij et al. (2006) and Smedes (2007). On average, absolute deviations between observed BAFs and those from these empirical relationships from Booij et al and Smedes were on average 0.24 and 0.32 log units for PAHs and PCBs in native mussels of station 2. Average absolute deviations of observed BAFs for transplanted mussels at station 1 and 2 were 0.40 and 0.35 log units and 0.32 and 0.41 log units, respectively, when comparing with regressions curves from Booij et al. and Smedes.

Elimination rate constants for PAHs and PCBs in transplanted blue mussels at Svensholmen are shown as a function of K_{ow} in Figure 9. The regression tool in Sigmaplot is used to obtain k_2. The k_2 values need to be treated with care as there is relatively large uncertainty in these values as shown by the

standard errors and P-values reported in Table 3 and Table 4. For PCBs, the k_2 values range between 0.016 d^{-1} for CB 52 to 0.043 d^{-1} for CB 101 in exposed blue mussels at Station 1 (Table 3); whereas for the PAHs, the k_2 values range between 0.008 d^{-1} for benzo[b,j]fluoranthene to 0.041 d^{-1} for fluoranthene in exposed blue mussels at Station 1 site (Table 4).

For Station 2, mussels of the final exposure period were lost, and therefore uptake curves for PAHs and PCBs with logKow > 5 did not present significant plateauing, and modelling with Equation 1 was difficult. Instead, we used an average of k_2 values from Station 1, C_m at t = 0 d and C_{Free} from passive samplers to predict C_{mussel} at 34 and 104 d. The relationship between predicted/observed concentrations in the deployed mussels at Station 2 and the hydrophobicity of the measured PAHs and PCBs is shown in Figure 10. In general, deviations between observed and predicted PAH and PCB concentrations in mussels are < 0.4 log unit, equivalent to no more than a factor of 2.5. Apart from the least hydrophobic PAH (naphthalene) there appear to be an increasing predicted/observed ratio with higher hydrophobicity.

4. Discussion

In the present study, non-contaminated blue mussels and passive sampler devices were deployed alongside native mussels within a moderately polluted city harbor area (Kristiansand, Norway) both to assess the contamination level at this location, compare the temporal pollutant bioaccumulation in transplanted vs. native mussels, and also to estimate bioaccumulation parameters contaminants using a first-order, single-compartment toxicokinetic approach.

At Station 1, the innermost harbor location, the finding of significantly increased nickel concentration in caged mussels corroborates recent monitoring at this site (Schøyen and Håvardstun, 2016) and is also supported by the fact that the site is in the vicinity of a nickel processing plant. According to data from caged mussels, Station 1 was generally more contaminated than Station 2, except for PAHs. Interestingly, Station 2 was located relatively close to the Fiskå bay area (Figure 1) which is home to an industrial company that earlier was known to be the main source of PAH pollution to the Kristiansand harbor waters. The finding was also strengthened by analyses of native mussels at Station 2, which showed elevated PAH levels as compared with typical background levels and with levels measured in the 0-group mussel. For metals, the analysis results in caged mussels indicated that a putative steady state was reached relatively fast, and generally faster than the non-polar, hydrophobic organic contaminants. A one-month deployment seemed to be long enough for stable concentration to be established. For the hydrophobic organic contaminants (PCBs and PAHs), a linear bioaccumulation occurred during the first months of caging, but the deployed mussels did in general not reach the concentrations detected in the native mussels, not even after six months of deployment. However, some of the least hydrophobic PCBs and PAHs (e.g. CB 28 and naphthalene) showed a different

pattern with a plateauing tendency already after one month, when contamination data on wet wt. basis were used, indicating the reaching of a steady state for these congeners. But this plateauing tendency was not seen for lipid-normalized concentrations.

The native mussel population at the Station 2 site was analyzed repeatedly during the six months' study period and these data clearly indicated the confounding influence from seasonal fluctuations on biological parameters (lipid %) as well as on chemical contaminant endpoints (especially PAHs but also some of the PCBs). In retrospect, repeated sampling and analyses of mussels from the donor-population (the Kaldvellfjord site) at each sampling date during the whole six months' study period would have provided a better basis for assessing the confounding influence from seasonal fluctuations in the present study. The PAH level in the native mussels decreased in the early phase of the study to a minimum during summer and then increased again towards the last sampling point in November. Many studies emphasize the relevance of considering seasonal fluctuations when interpreting contaminant data (and also biomarker signals) in blue mussels, e.g. (Björk and Gilek, 1997; Westerbom et al., 2002; Orban et al., 2002; Pfeifer et al., 2005; Leinio and Lehtonen, 2005; Nesto et al., 2007; Farcy et al., 2013; Schmidt et al., 2013; Mugica et al., 2015). The annual cycle will in a complex and dynamic manner influence pollutant bioaccumulation processes that occur *in situ* and the biological condition and pollution responses that appear *in vivo* in mussels. In this regard, variability in nutritional/growth and reproductive (e.g. spawning and gametogenesis) factors are important, regardless of whether the endpoints of the study are chemical exposure markers or ecotoxicological effect markers. Nevertheless, the mussel sentinels deployed in the Kristiansand harbor in the present study, rapidly changed from the pre-deployment level by accumulating increased levels of ecotoxicologically relevant metals, PAHs and PCBs, although most of the PAHs showed consistently lower concentrations than the levels found in the native mussel collected from the Svensholmen site. However, it may be argued that steady-state conditions may not need to be reached for deployed mussels if the objectives of the study are to compare contamination levels at different field sites or for establishing time trends, so long as the kinetics of accumulation are the same at all sites (i.e. same time of year, similar water temperature, same deployment design, etc.). The generally higher PAH levels in native mussels than in transplanted mussels in the present study is most likely because the native mussels had a much longer time of exposure (several years). The long-term bioaccumulation of PAHs in native mussels is a complex process and the modelling of this process requires the use of multi-compartment uptake and elimination modelling tools, e.g. (Stegeman and Teal, 1973), or toxicokinetic models such as those based on dynamic energy budget (DEB) theory, e.g. (Vanharen and Kooijman, 1993; Vanharen et al., 1994), which can estimate the dynamic influence on bioaccumulation by multiple factors related both to the pollutant, the environment, and the physiological condition of the sentinel organism.

However, first-order, single-compartment models are still the most common tools in the ecotoxicological studies for studies of contaminant bioaccumulation and for estimating BAFs. So, what could be the best endpoint for indicating that steady-state contaminant concentrations are obtained in deployed mussels: an observed plateauing of C_m/C_{free} or the determination of identical C_m in native and transplanted mussels? In our study, there was apparently a systematic trend towards a higher predicted/observed ratio with higher K_{ow} values within both the PCBs and PAHs chemical classes (Figure 10). Other studies have found that the linear relationship between logBAF and logK_{ow} does not hold for compounds with logK_{ow} > 6. Devillers et al. (1996), and Barthe et al. (2008) suggested that this phenomenon is caused by steric hindrance of permeation through biological membranes by the larger (and higher K_{ow}) contaminant molecules. Here, the accumulation and depuration rates were different for PAHs and PCBs. The slope of the regression of logBAF against logK_{ow} was slightly lower than 1 (0.92 for PCBs and 0.76 for PAHs) (Table 5), which is similar to that reported by Booij et al. (2006) in a review study that addressed the three mussel species *Mytilus edulis*, *M. complanata*, and *Perna viridis*. The intercepts in our study are apparently a bit lower than those reported by Booij et al. (2006) and it should be noted that the reported ratios span 4 orders of magnitude. The BAF values obtained in this study are consistent with the values by the Booij et al. (2006) review. As expected from contaminant masses found in native and transplanted mussels, differences in BAFs can be seen for native and transplanted mussels.

The time required for deployed mussels to reach steady state for different contaminants will obviously depend on the toxicokinetic properties of the specific pollutant substance, on factors attributed to the mussel (condition, reproductive state, etc.), as well as on recipient factors *in situ*. While the uptake of hydrophobic (non-polar) organic contaminants, such as PCBs and PAHs, occur as a passive diffusive process/equilibrium partitioning, other and more complex mechanisms are thought to be involved for trace metals, as their accumulation by mussel and other aquatic organisms is influenced by a variety of factors, such as multiple routes of exposure (diet and solution), metal speciation, ligand associations and complexation, chemical composition of the surrounding medium and physiological or biochemical effects on bioavailability (Luoma, 1983; Simkiss and Taylor, 1989; Luoma and Rainbow, 2005). According to Jenne (1977), the bioavailability of trace metals to mussels may be influenced by at least four factors: (1) the physiological and ecological characteristics of the mussels, (2) the forms of dissolved elements, (3) the forms of elements in ingested solids, and (4) the chemical and physiological characteristics of the seawater. For instance, Cd entry into cells of gills of marine mollusks may occur through calcium channels (Roesijadi and Unger, 1993). Some elements are also essential, meaning that they are necessary for optimal growth, development and homeostasis. As such, organisms are capable of regulating these metals (Lobel and Marshall, 1988). Differences in accumulation between transplanted and native mussels may therefore be a consequence of physiological differences (e.g. pertaining to their general condition), resulting in different uptake

and/or depuration rates for specific elements. This could be related e.g. to different ingestion rates, different transport through ion channels at the cell membrane, or different concentrations of metal-binding proteins, such as metallothionein. Cd may e.g. compete with Ca for transmembrane transport by calcium pump in the epithelia of mussels. The rapid bioaccumulation of trace metals in caged mussels in the present study, in particular at Station 1, has also been observed by others. Regoli and Orlando (1994) studied the uptake of Pb, Fe, and Mn in *Mytilus galloprovincialis* deployed at a metal polluted site and reported that a steady state was reached after only 2 weeks, suggesting that mussel rapidly equilibrate with the elevated environmental levels of metal pollutants. It can be expected that native and transplanted mussels experience exposure to the same forms/species of the elements (dissolved or associated with solids) and water characteristics, however, these may change with time, and thus explain some of the differences observed between mussels sampled in July (34 days) and mussels sampled in September (104 days). With respect to PCBs and PAHs, there was a good correspondence in the present study between levels determined in mussels and levels determined in water through the use of passive samplers. Moreover, the observed bioaccumulation in mussels revealed only little systematic difference between PCBs and PAHs (e.g. Figure 8, Figure 9, and Figure 10). The correspondence between mussels and passive samplers for PCBs and PAHs was actually better than expected, especially for the PAHs which in these industrially influenced harbor waters can be expected to be predominantly associated with microscopic coal tar pitch and soot particles of a local industrial origin. Such particles may contain PAHs both adsorbed to the particle surface as well as absorbed in the internal particle matrix, and they are key factors for the partitioning, bioavailability, uptake, and bioaccumulation of PAHs in aquatic environments (Gustafsson and Gschwend, 1997; Gustafsson et al., 1997; Cornelissen et al., 2005). In the present study, PAHs from a predominately soot particle origin would be expected to be less available for uptake by passive samplers in comparison to by the mussels, as the active filter feeding process of mussels will provide more routes for uptake (both from diet and solution). Importantly, as the mussels in this study were not depurated prior to analysis, PAH associated to particles contained in the mussel gut must have contributed to the detected concentration level. However, the observed high degree of correspondence between PCB and PAH accumulation in the mussels supports an assumption that passive uptake from the solved fraction was the dominating uptake route for both contaminant classes. And this uptake route is also what is measured by the passive samplers.

In summary, a general increase of contaminant loads was recorded when clean blue mussels were deployed in the waters of the Kristiansand harbor. The increase was most significant for certain metals (Ni and Cu) at Station 1 (the industrial site), which is in agreement with our previous monitoring data of this site. At Station 2, the measured PAH levels (in both caged and native mussels) were markedly higher than at the Station 1, which most likely was due to the closer proximity of Station 2 site to another industrialized site which historically was (and probably still is) the main source for PAH

contamination to the Kristiansand harbor waters. The native mussels present at Station 2 were significantly more contaminated by PAHs and PCBs than the mussels deployed in the harbor study area, also after six months' deployment. The high K_{ow} PAHs displayed a higher native - transplant ratio than the trace metals, indicating that a longer time than six months is required for steady state to establish. In order to establish steady state for all monitoring-relevant contaminants, the caged mussels will probably need to be deployed for exposure times that are in the range of the age of native mussels. Although this is possible to achieve, it will not be practically feasible for most mussel monitoring studies. Rather, from a practical viewpoint, short mussel deployments are by most means desirable as it reduces both field costs as well as the risk of practical problems such as biofouling or the loss of caging rigs. Our results suggest that the optimal deployment time in mussel caging is highly substance dependent, with the most hydrophobic organic contaminants requiring very long deployments. Short term mussel deployments, such as one-two months, appear to be suitable for trace metals and the less hydrophobic non-polar organic contaminants. However, such quite short deployments may also work fine for monitoring of other, more hydrophobic, organic contaminants as long as the study aim is to compare time trends and relative exposure loads at different field sites, and not to describe contaminant concentrations under real steady state conditions. For the latter issue, the use of very long mussel deployments would be recommended, if the collection and analysis of native mussel sentinels is not possible at the study site. Otherwise, a toxicokinetic modelling approach must be used to predict theoretical steady state levels from contaminant concentration data in blue mussels that have been deployed only for short periods of time or from passive samplers, or both.

Acknowledgments

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542

544 Legends of figures:

545 **Figure 1. Localization of the two mussel stations 1 and 2 (industrial site and Svensholmen) in the harbor area of the**
546 **Kristiansand city, southern Norway.**

547 **Figure 2. Temporal changes in trace element concentrations in transplanted blue mussels (mg kg^{-1} wet wt.) exposed at**
548 **Station 1 (industrial site) (○) and at Station 2 (Svensholmen) (□) for deployment times of 34, 104 and 181 days. Note**
549 **the different scales on the y-axes.**

550 **Figure 3. Concentrations of trace elements (mg kg^{-1} wet wt.) in transplanted blue mussels at Station 1 (industrial site)**
551 **and Station 2 (Svensholmen) (after 34, 104 and 181 days of exposure) and in native blue mussels (Station 2 only;**
552 **sampled on 4 occasions) as a function of DGT-labile concentrations (µg L^{-1}).**

553 **Figure 4. Ratio of trace element concentrations in transplanted blue mussels over those in native blue mussels at**
554 **Station 2 sampled on July 2nd (●) and Sept. 10th 2015 (○). Error bars are calculated from relative percent deviations**
555 **of duplicate analyses for transplanted and native mussels.**

556 **Figure 5. PCB accumulation curves in transplanted blue mussels exposed at Station 1 (industrial site). The y-axis**
557 **C_m/C_{free} (L kg^{-1}) represents the concentration in mussels (C_m in ng g^{-1} wet wt.) over the freely dissolved concentration**
558 **in water (ng L^{-1}) determined by passive sampling for each sampling time (0, 34, 104 and 181 days). Duplicate pooled**
559 **mussel samples were analyzed. See material and methods section (section 2.3) and Equation 1 for model lines.**

560 **Figure 6. Accumulation curves over time for PCBs on wet (w.) and lipid (l.) wt. basis in transplanted mussels at**
561 **Station 1 (industrial site), and both transplanted and native mussels at Station 2 (Svensholmen). Curve fitting was for**
562 **visual impression and has no mathematical meaning.**

563 **Figure 7. Accumulation curves over time for PAHs on wet (w.) and lipid (l.) wt. basis in transplanted mussels at**
564 **Station 1 (industrial site), and both transplanted and native mussels at Station 2 (Svensholmen). Curve fitting was for**
565 **visual impression and has no mathematical meaning.**

566 **Figure 8. Logarithm of bioaccumulation factor (log BAF, calculated as the contaminant concentration wet wt. in blue**
567 **mussels over the freely dissolved concentration) for PAHs (empty symbols) and PCBs (filled symbols) for native**
568 **mussels at Station 2 (Svensholmen), transplanted mussels at Station 2 and transplanted mussels at Station 1**
569 **(industrial site) as a function of $logK_{ow}$. See the text for derivation of log BAF values. Error bars for log BAF for**
570 **native mussels from Station 2 represent standard deviation calculated from log BAF estimated at time 0, 34, 104, and**
571 **181 days of the experiment (n = 4). The solid and dashed lines represent logBAF-logKow regressions from Booij et al.**
572 **(2006) (log BAF = 0.84 $logK_{ow}$ -0.49) and Smedes (2007) (log BAF = 1.1 $logK_{ow}$ -2.14), respectively.**

573 **Figure 9. First-order depuration rate constants, k_2 for PAHs and PCBs in transplanted blue mussels exposed at**
574 **Station 2 (Svensholmen). The regression shown is from Booij et al. (2006).**

575 **Figure 10. Logarithm of the ratio of predicted PCB and PAH concentrations as a function of K_{OW} in blue mussels over**
576 **observed concentrations for transplanted blue mussel exposures of 34 and 104 days at Station 2 (Svensholmen).**

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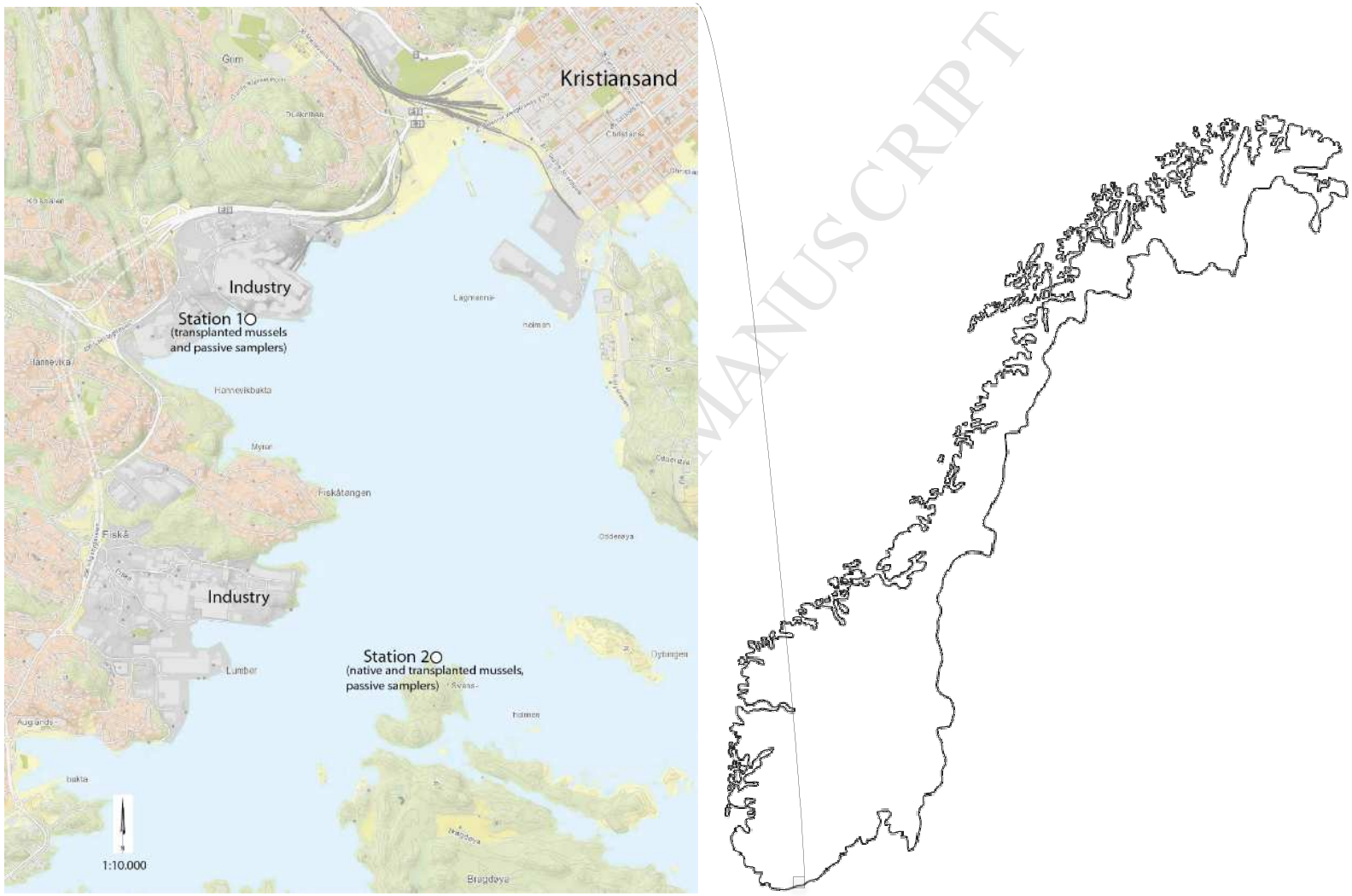
579 Figure 1



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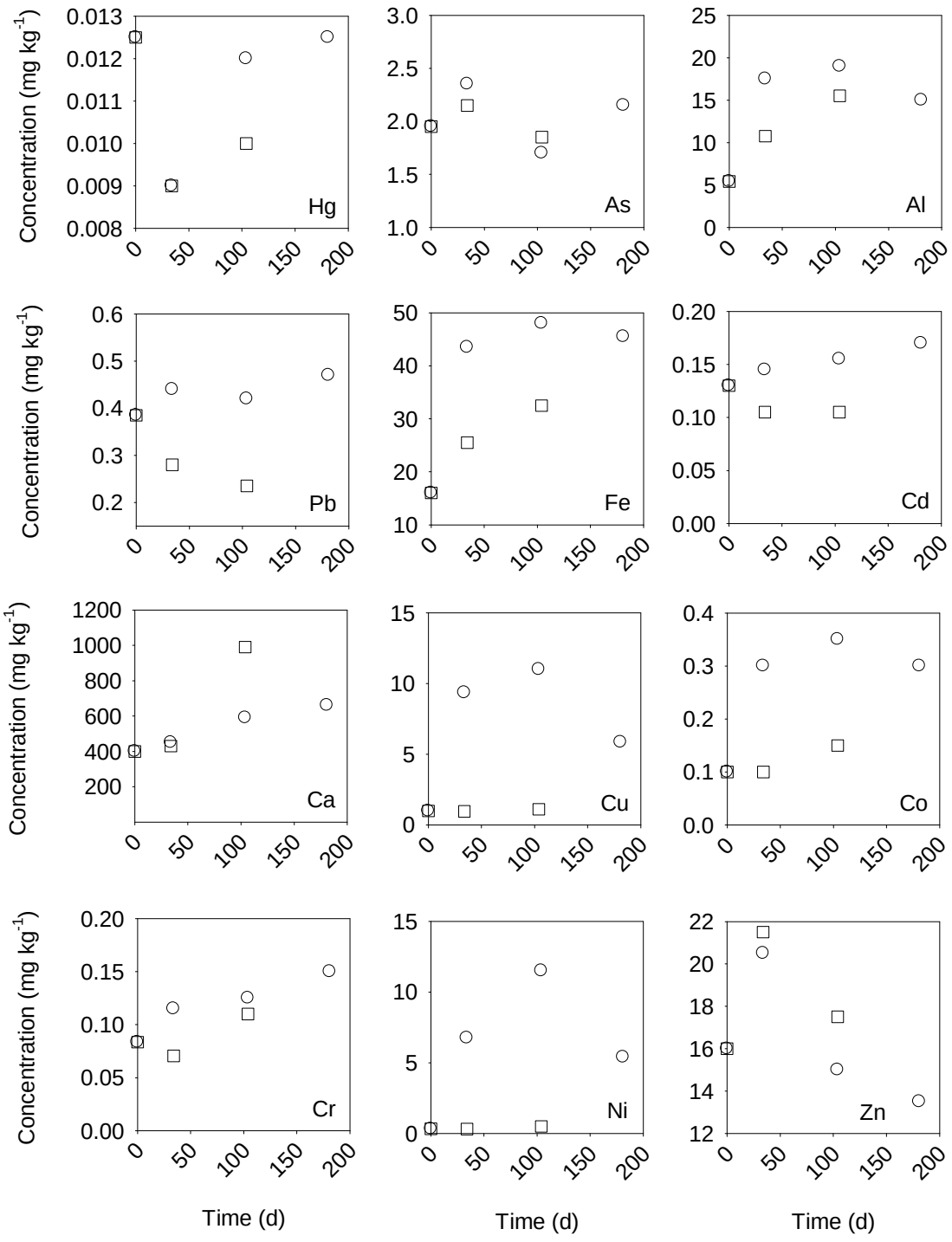
582 Figure 2



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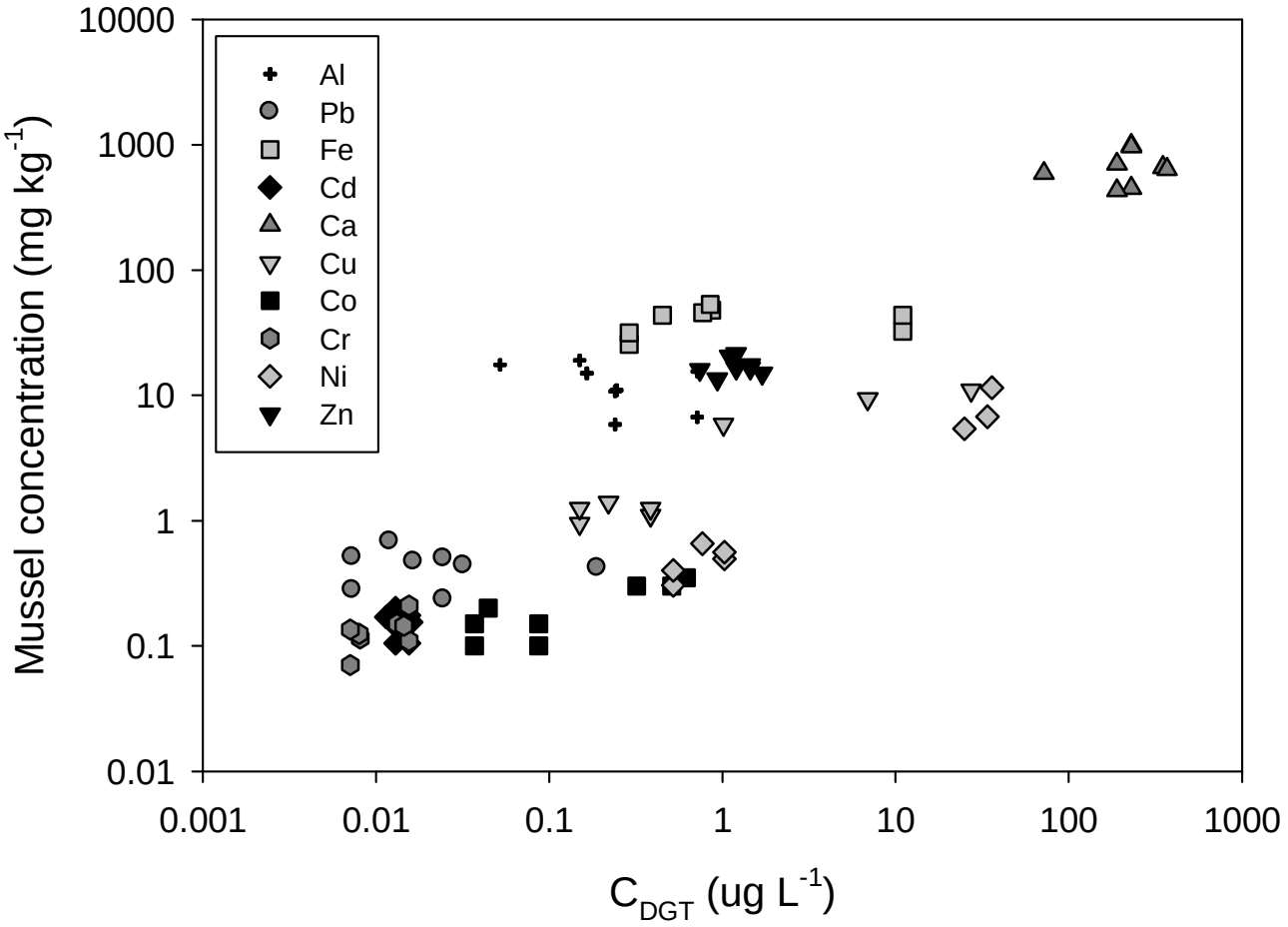
585 Figure 3



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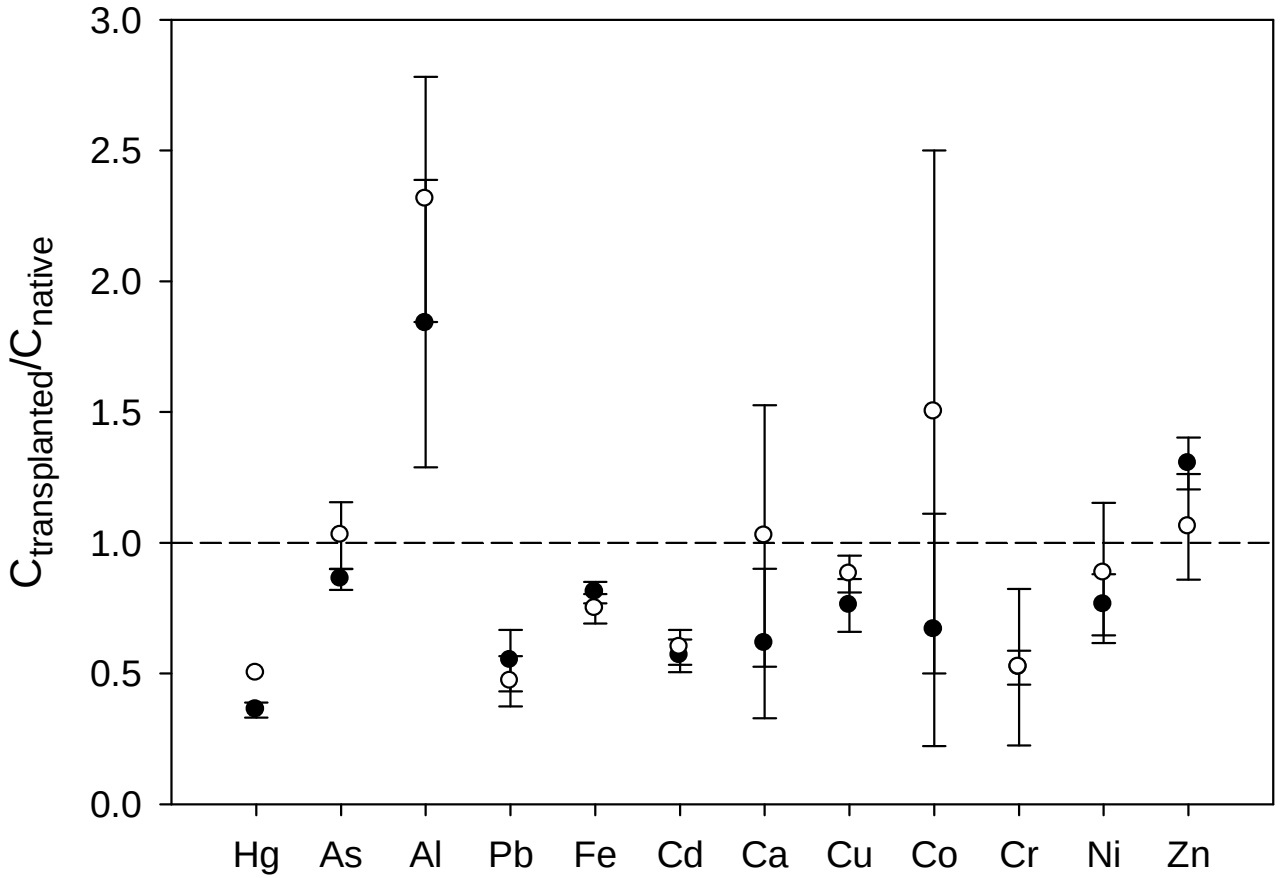
588 Figure 4



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591 Figure 5

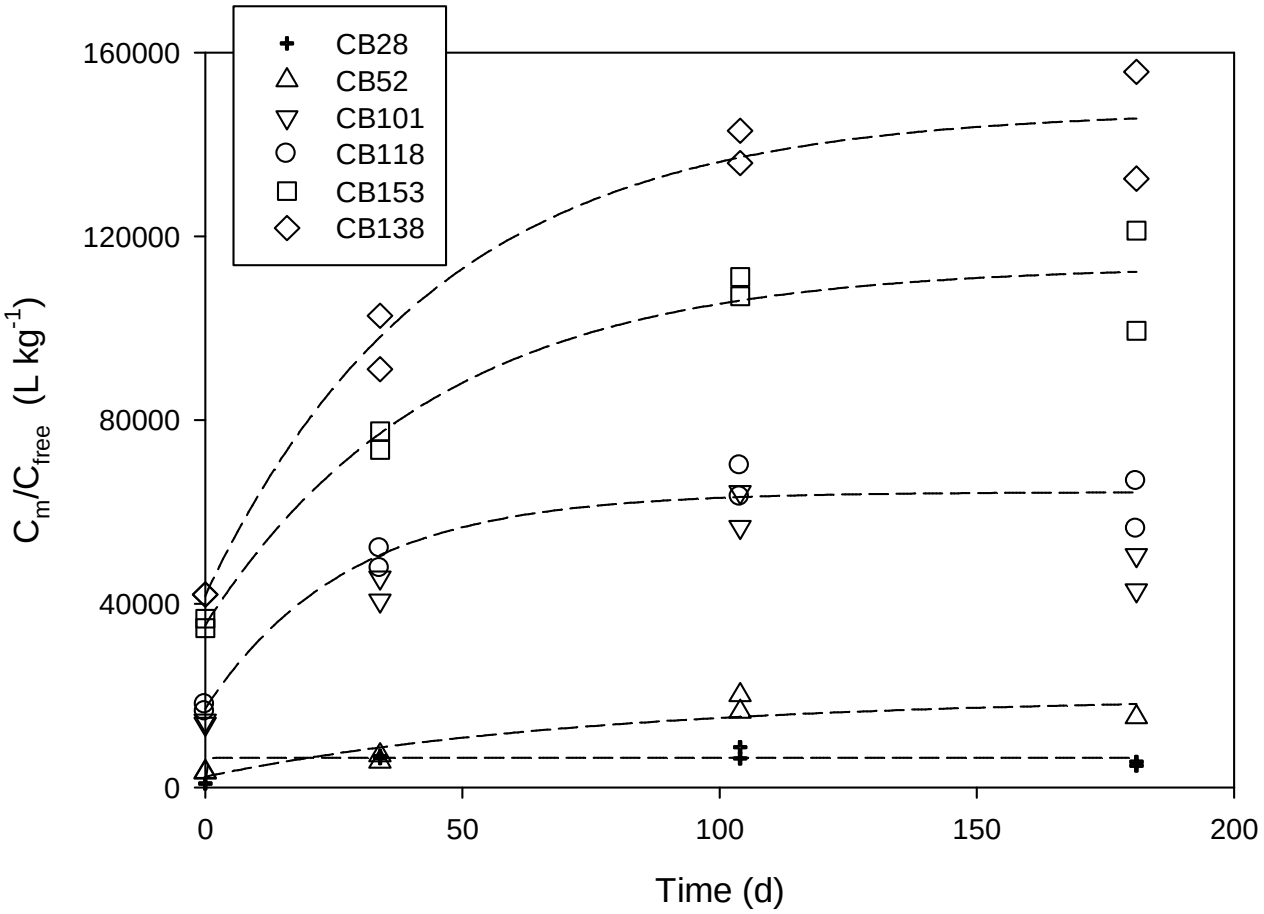

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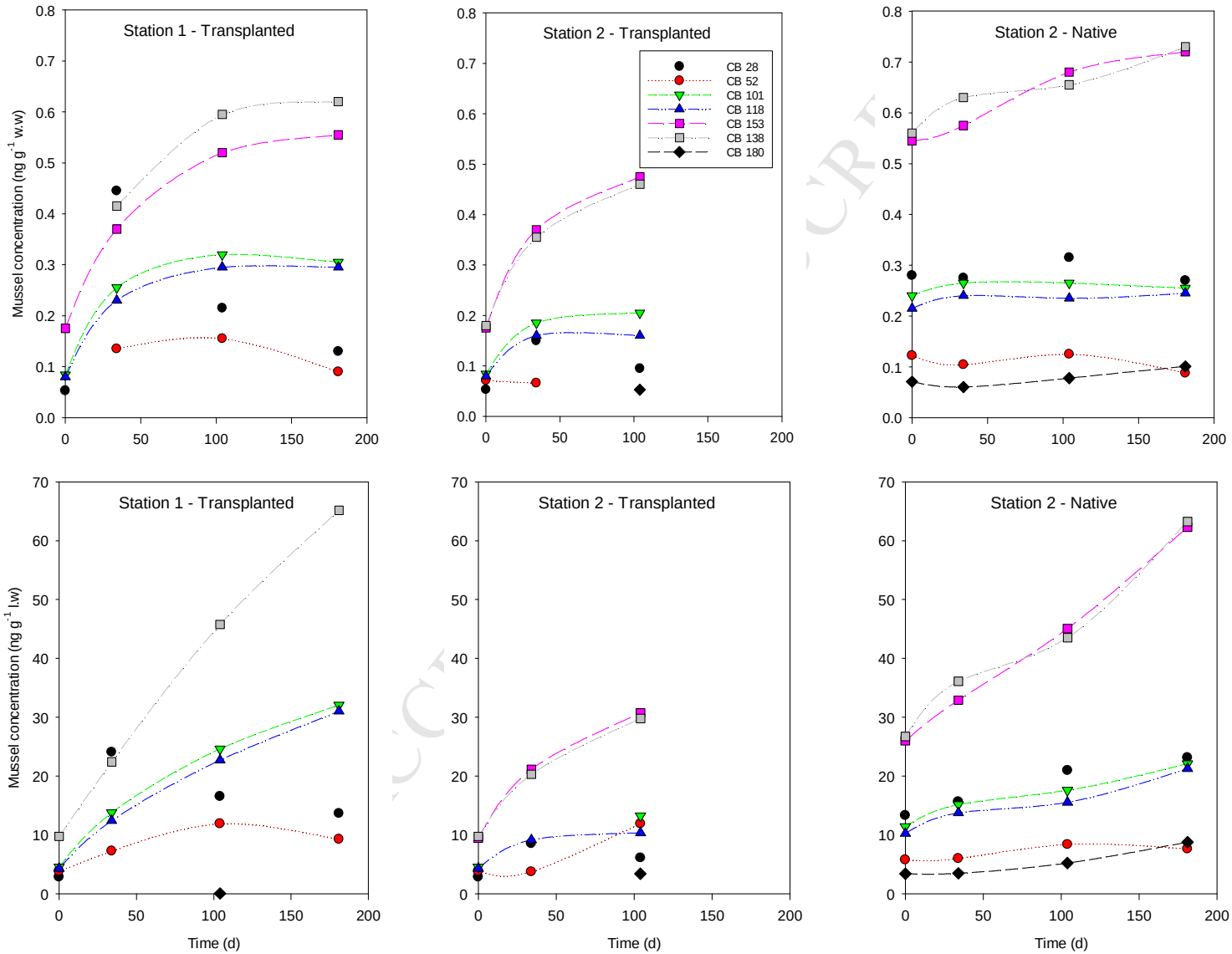
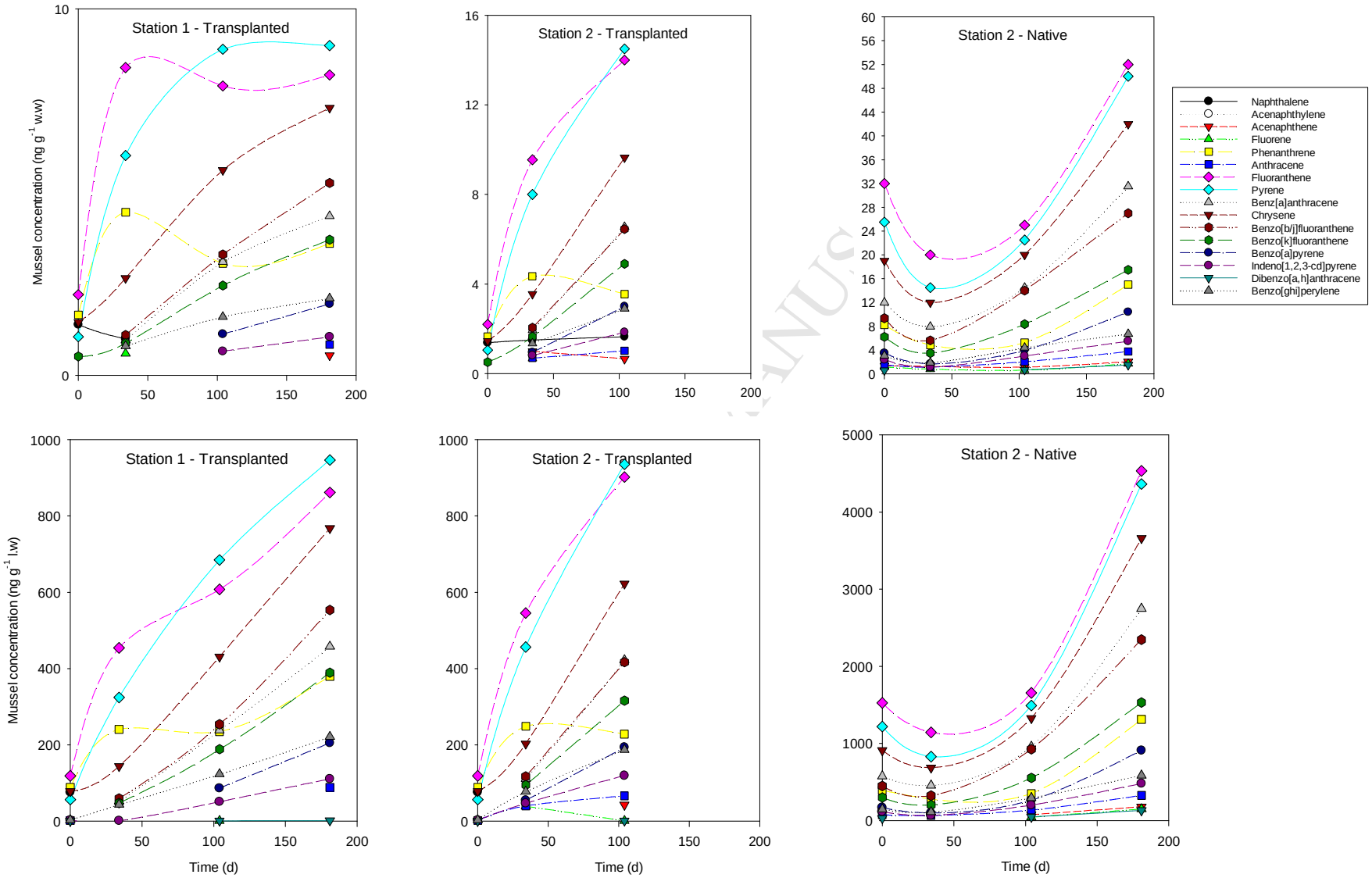
Figure 6


Station 1 - Transplanted
Station 2 - Transplanted
Station 2 - Native
CB 28
CB 52
CB 101
CB 118
CB 153
CB 138
CB 180
Mussel concentration (ng g-1 w.w)
Mussel concentration (ng g-1 l.w)
Time (d)

597 Figure 7



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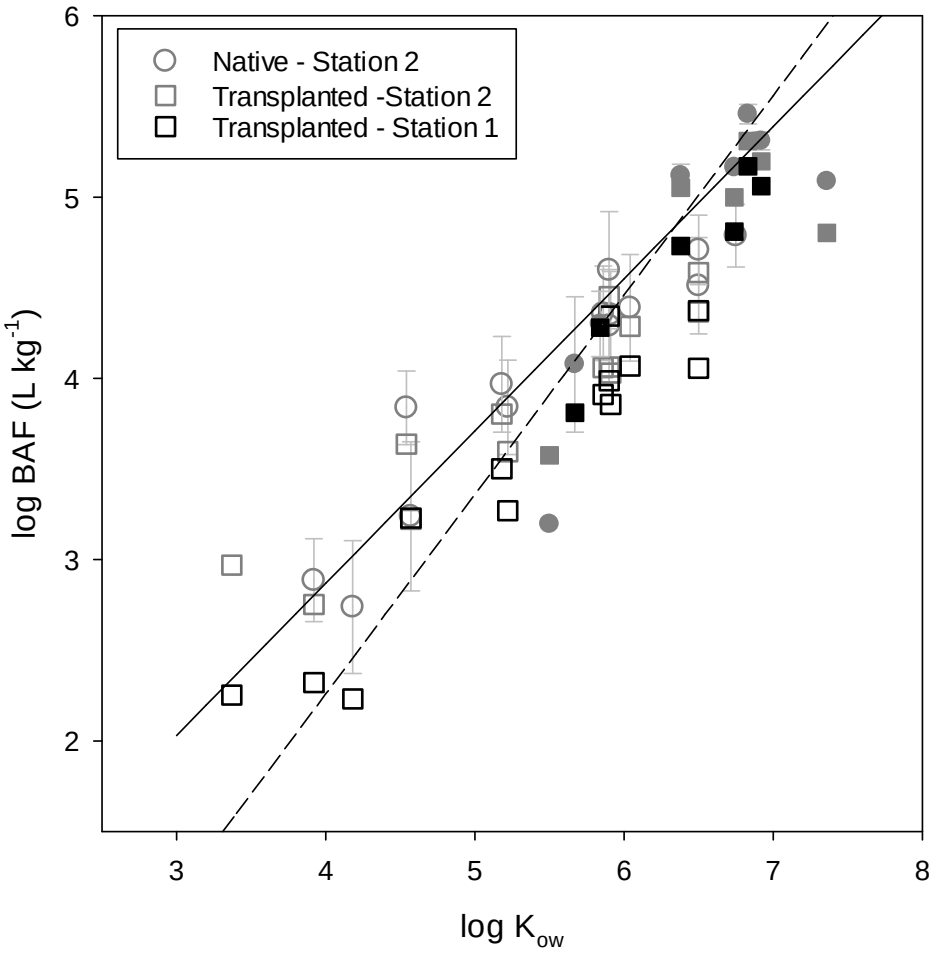
601 Figure 8



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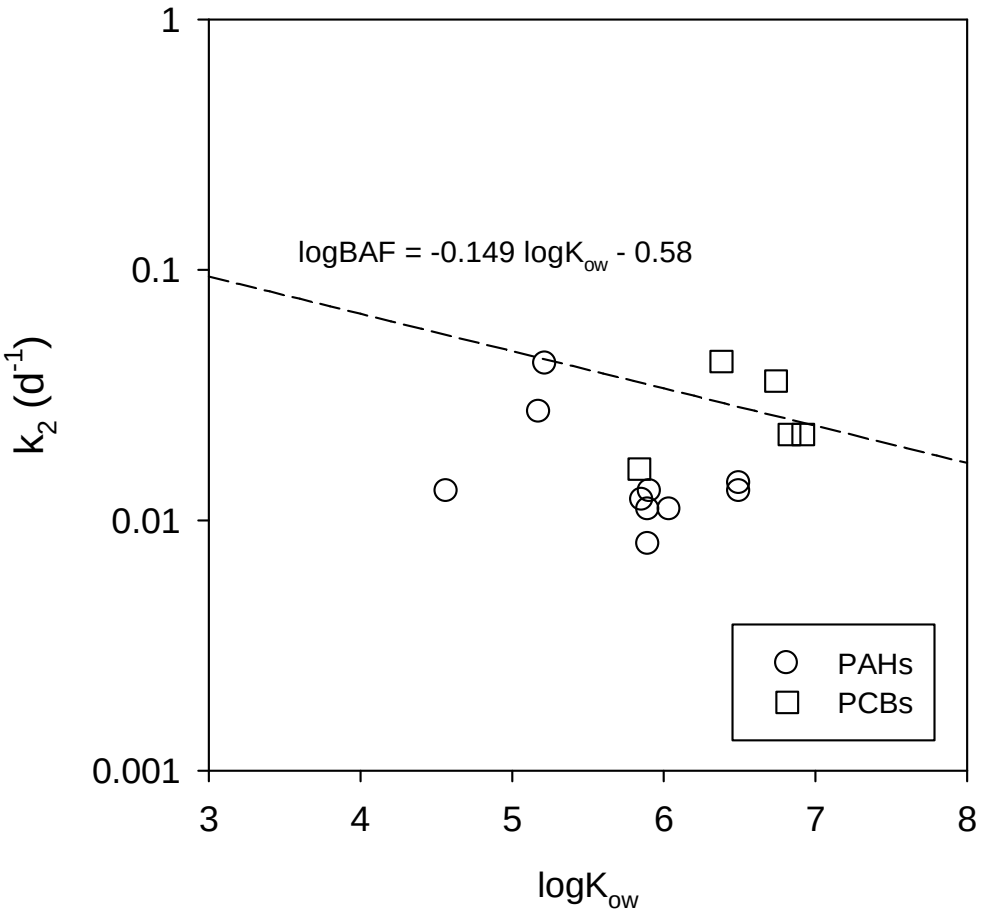
604 Figure 9



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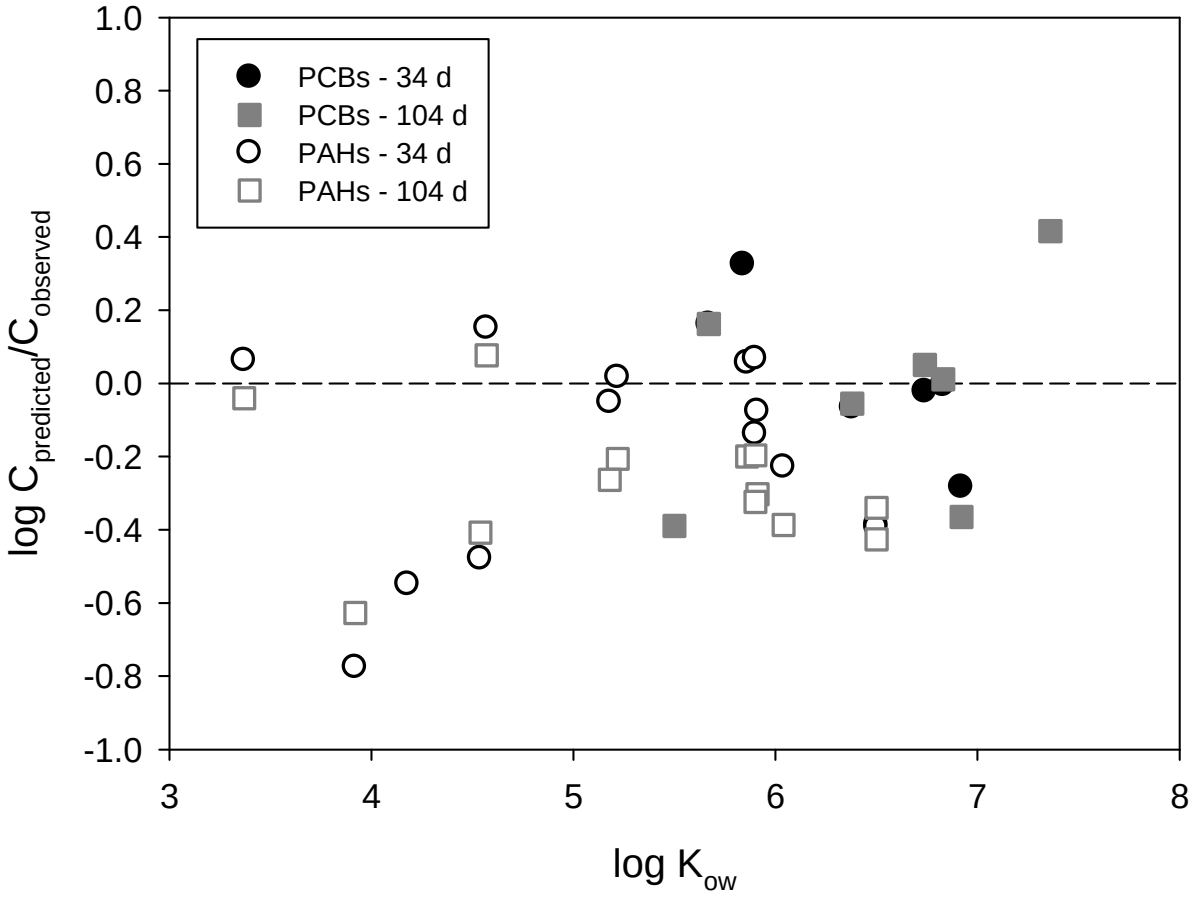
607 Figure 10



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612 **Table 1: Contaminant concentrations (wet wt.) in deployed and native blue mussels in the Kristiansand harbor area at Station 1 (industrial site) and Station 2 (Svensholmen). All**
613 **concentrations are shown by the mean concentration of two replicate composite samples. For comparison, the rightmost columns show the 10 and 90 percentile concentration levels in**
614 ***M. edulis* from background and slightly impacted stations in the Norwegian coastal monitoring program, see Beyer et al. (this volume) for more details. Data shown in shaded boxes**
615 **are above the 90 percentile level.**

616

617 (Table 1 is uploaded separately as an excel file)

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622 **Table 2. Blue mussel-water bioaccumulation factors (log BAF, wet wt.; L kg^{-1}) for trace elements measured in transplanted and native mussels (mg kg^{-1} wet wt.) at two exposure sites**
623 **based on DGT-labile concentrations in water (C_{DGT}; µg L^{-1}).**

	Mussel-water bioaccumulation factor (log BAF; L kg^{-1})							
	Station 1 (industrial site)			Station 2 (Svensholmen)				
	Transplanted			Transplanted		Native		
	34 d	104 d	181 d	34 d	104 d	July 2nd	Sept. 10th	Nov. 26th
Al	5.53	5.10	4.96	4.65	4.34	4.39	3.97	4.65
Pb	4.14	3.34	4.45	4.58	3.98	4.84	4.31	4.76
Fe	4.99	4.74	4.77	4.94	3.47	5.04	3.60	4.79
Cd	4.05	3.99	4.17	3.91	3.83	4.15	4.05	4.19
Ca	3.29	3.91	3.28	3.35	3.63	3.57	3.62	3.23
Cu	3.13	2.61	3.76	3.80	3.46	3.92	3.51	3.80
Co	2.77	2.75	2.97	3.43	3.24	3.61	3.06	3.65
Cr	4.15	4.19	4.06	4.00	3.85	4.28	4.13	4.00
Ni	2.30	2.50	2.33	2.77	2.68	2.89	2.74	2.93
Zn	4.27	3.95	4.16	4.25	4.08	4.14	4.06	4.33

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627 **Table 3. Results from two parameter modelling of the uptake of PCBs into blue mussels exposed at Station 1 (industrial site).**

Parameters	BAF (L kg^{-1})*			Depuration rate, k_2 (d^{-1})			R^2	log BAF	$t_{90\%}$ (d)**
	BAF	SE	P-value	k_2	SE	*p*-value			
PCB31+28	6420		< 0.0001					3.81	
CB52	19139	4568	0.0086	0.016	0.01	0.153	0.80	4.28	144
CB101	53431	4004	< 0.0001	0.043	0.021	0.0930	0.88	4.73	54
CB118	64318	2888	< 0.0001	0.036	0.01	0.0141	0.96	4.81	64
CB153	113580	5708	< 0.0001	0.023	0.006	0.0120	0.96	5.06	105
CB138	147551	8398	<0.0001	0.022	0.005	0.0066	0.97	5.17	105

$$\frac{C_m}{C_{Free}} = Y_0 + (BAF - Y_0)(1 - e^{-k_2 t})$$

with $Y_0 = C_{m(t = 0)}/C_{free\ (34\ d)}$

*wet wt.-based bioaccumulation factors

**$t_{90\%}$ calculation based on the estimated depuration constant k_2 and that the mussel concentration for the contaminant of interest is negligible

SE: standard error

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631 **Table 4. Results from two-parameter modelling of the uptake of PAHs into blue mussels exposed at Station 1 (industrial site).**

Parameters	BAF (L kg^{-1})*			Depuration rate, k_2 (d^{-1})			R^2	log BAF	$t_{90\%}$ (d)**
	BAF	SE	p-value	k_2	SE	*p*-value			
Phenanthrene	1702	418	0.0097	0.012	0.008	0.2016	0.881	3.23	177
Fluoranthene	1854	158	< 0.0001	0.041	0.021	0.1144	0.872	3.27	54
Pyrene	3196	408	0.0005	0.027	0.015	0.1253	0.85	3.50	85
Benz[a]anthracene	7182	1663	0.0125	0.013	0.007	0.1523	0.803	3.86	177
Chrysene	8187	1708	0.0049	0.012	0.007	0.1601	0.909	3.91	192
Benzo[b/j]fluoranthene	9686	2868	0.0278	0.008	0.005	0.1445	0.899	3.99	288
Benzo[k]fluoranthene	22081	5772	0.0123	0.011	0.006	0.1323	0.871	4.34	209
Benzo[a]pyrene	11682	3468	0.078	0.011	0.009	0.3122	0.492	4.07	209
Indeno[1,2,3-cd]pyrene	11355	2897	0.0594	0.014	0.011	0.3298	0.35	4.06	164
Benzo[ghi]perylene	23660	2927	0.0013	0.013	0.004	0.0301	0.92	4.37	177

$$\frac{C_m}{C_{Free}} = Y_0 + (BAF - Y_0)(1 - e^{-k_2 t})$$

with $Y_0 = C_{m(t = 0)}/C_{free\ (34\ d)}$

*wet wt.-based bioaccumulation factors

**$t_{90\%}$ calculation based on the estimated depuration constant k_2 and that the mussel concentration for the contaminant of interest is negligible

SE: standard error

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635 **Table 5. Results of the linear regression of log BAF with logK_{ow} for PAHs and PCBs for native (Station 2, Svensholmen) and transplanted (Station 2, Svensholmen and Station 1,**
636 **industrial site) mussels.**

			n	Slope			Intercept			R^2
				a	SE	*p*-value	Y_0	SE	*p*-value	
PAHs	N	Station 2	14	0.692	0.066	< 0.0001	0.23	0.37	0.54	0.90
	T	Station 2	14	0.554	0.057	< 0.0001	0.88	0.31	0.016	0.90
	T	Station 1	13	0.738	0.069	< 0.0001	-0.43	0.37	0.27	0.92
PCBs	N	Station 2	8	1.02	0.24	0.0051	-1.8	1.5	0.28	0.71
	T	Station 2	7	0.86	0.25	0.018	-0.9	1.6	0.60	0.71
	T	Station 1	6	0.924	0.134	0.0023	-1.3	0.9	0.22	0.92
$\log BAF = a\ logK_{ow} + Y_0$ N: native mussels; T: transplanted mussels; SE: standard error										

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MERE_2017_201

Highlights

- Bioaccumulation of anthropogenic contaminants in deployed and native blue mussels during a six-month period was studied.
- Bioaccumulation factors for metals and organic contaminants were estimated.
- Differences in contaminant levels in transplanted and native mussels were observed.
- Significant confounding influence from seasonal factors on contaminant concentrations was observed.
- Standardization and harmonization of monitoring techniques that involve deployed and native blue mussels are needed.